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(54) Method of manufacturing polybutylene terephthalate resin films.

(57) A polybutylene terephthalate resin film is produced by turning into film a polybutylene terephthalate resin having an inherent viscosity of more than 1.0 by the inflation molding technique and under the condition of extrusion resin temperatures within a range defined by the following relation:

melting point (°C) < extrusion resin temperature (°C) < melting point - 26 + 53 x inherent viscosity (°C)

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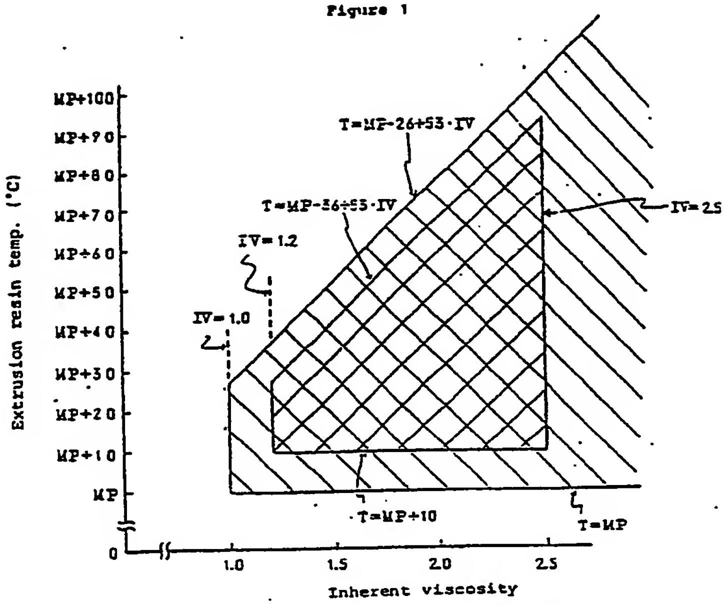
Figure 1



Practicable range: temp. range which satisfies the relation:
MP < T < MP − 26 + 53·IV, 1.0 < IV

Preferred range: temp. range which satisfies the relation:
MP + 10 ≤ T ≤ MP − 36 + 53 · IV, 1.2 ≤ IV ≤ 2.5

where MP: melting point;
T: extrusion resin temp.;
IV: inherent viscosity

METHOD OF MANUFACTURING POLYBUTYLENE TEREPHTHALATE RESIN FILMS

(Field of the Invention)

This invention relates to a novel method of making polybutylene terephthalate resins film.

(Brief Description of the Drawing)

Figure 1 is a graph showing extrusion resin temperature ranges applicable to inflation molding according to the method of the invention.

(Prior Art)

Polybutylene terephthalate resins are widely used as typical engineering plastics.

Most of these resins, however, are exclusively used for injection moldings, and are rarely used for extrusion moldings, and films in particular. The reason for this is that molding such resin into films is extremely difficult.

Generally, there are two types of methods for production of films, namely, T-die method and inflation method. The T-die method is usually such that a stream of molten plastics introduced through one inlet is broadened to a desired film width and made as uniform in thickness as possible, so that the melt in extruded in a thin film pattern through a slit-shaped nozzle, the extruded resin being then cooled into a film. The inflation method, often employed in molding polyethylene into film, comprises extruding molten plastic through an annular extrusion nozzle to form a tube, the tube being inflated by blowing a fluid, e.g., air, thereinto, whereby the extruded resin is formed into a tubular film. The T-die and inflation methods have their respective advantages and disadvantages. Generally, the inflation method, as compared with the T-die method, provides higher productivity and greater economy and is suitable for production of thin films, but its applicability varies according to the kind of resin material used. As such, it cannot be said that the method is applicable for use with any kind of resin, and naturally the range of resin varieties with which the method can be employed is limited to a few special kinds of resins, such as polyolefins. Generally, polyesters, such as polyethylene terephthalate and polybutylene terephthalate, are film-moldable by the T-die method, but on the other hand, manufacturing of films of these resins involve various technical problems to be solved and, as such, it has not been industrialized to date.

In view of this situation, the present inventors conducted a series of studies directed to making it possible to produce films of polybutylene terephthalate by the inflation technique, and as a result, they found that polybutylene terephthalate could be adapted for film extrusion by the inflation technique by selecting a specific variety of polybutylene terephthalate and a specific range of temperature conditions. This discovery led to the present invention.

(Summary of the Invention)

Accordingly, this invention provides a method of manufacturing a polybutylene terephthalate resin film characterised in that a polybutylene terephthalate resin having an inherent viscosity of more than 1.0 is turned into film by an extrusion inflation molding technique within an extrusion resin temperature range defined by the following relation:

melting point (°C) < extrusion resin
temperature (°C) < melting point
- 26 + 53 x inherent viscosity - (°C)

The invention also includes within its scope polyethylene terephthalate resin film whenever prepared by a method as defined above.

The term "inherent viscosity" used herein refers to a value therefor as measured in orthochlorophenol at 25°C.

After their series of studies with polyakylene terephthalate resins, the present inventors found that whereas stable film making of polyethylene terephthalate resins by inflation molding is extremely difficult under any conditions, it is possible to filmize a polybutylene terephthalate, only if its inherent viscosity (hereinafter referred to as IV) is more than 1.0, by inflation-molding same in such a way that the extrusion resin temperature comes within the range covered by the foregoing relation.

Such polybutylene terephthalate resin of more than 1.0 in IV is higher in IV and greater in average molecular weight than ordinary-types of polybutylene terephthalates, and those having an IV of 1.2 - 2.5, inter alia, are preferred, which may be in the form of either a homopolymer or a copolymer consisting principally of a polybutylene terephthalate. One having an IV of more than 2.5 can hardly be produced as such. One having an IV of less than 1.0 is undesirable because it is difficult to filmize because of drawdown possibilities during inflation molding. However, a mixture of a polybutylene terephthalate having an IV of less than 1.0 and one having an IV of more than 1.0 is filmizable if the resulting IV of the mixture is more than 1.0.

For the purpose of inflation molding, it is essential that in melt extruding through an annular nozzle a polybutylene terephthalate resin having such a specific IV value as above defined by the inflation technique, the temperature of the resin should be within a range defined by the following relation:

melting point (°C) < extrusion resin
temperature (°C) < melting point
- 26 + 53 x inherent viscosity - (°C)

The polybutylene terephthalate resin used is preferably of an IV of 1.2 - 2.5, and especially preferably of an IV of 1.5 - 2.2, and the temperature of the resin is preferably within a range defined by the following relation;

melting point + 10 (°C) < extrusion
resin temperature (°C) < melting point
- 36 + 53 x inherent viscosity (°C)

These ranges of resin temperatures for inflation molding are shown in Figure 1, which is a graph of IV plotted against extrusion temperature. The area shown with single hatching is the practicable range, which satisfies the conditions : melting point < extrusion resin temperature < melting point - 26 + 53 Iy, and IV > 1. The area shown with double hatching is the preferred range which satisfies the conditions: melting point ≤ extrusion resin temperature ≤ melting point - 36 + 53 IV, and 1.2 ≤ IV ≤ 2.5. If the resin temperature is too high, the viscosity of the melt becomes so low that making film from the resin is impracticable because of drawdown possibilities. If the temperature is too low, the viscosity of the melt becomes so high that grerater pressure loss in the die is involved, the power requirements becoming greater for operation of the extruding machine, which is followed by decreased productivity. Another undesirable effect of such low temperature is increased film thickness irregularity due to non-uniformity of resin flow in the die.

According to the invention, film making of a polybutylene terephthalate resin by the inflation technique is possible by maintaining the foregoing essential requirements, and for the other conditions, general conditions of the inflation technique are applicable. By employing a crosshead die, a tubular melt of polybutylene terephthalate resin is extruded upward or downward; the tube is held at one end between the pinch rolls and air is fed thereinto, so that the tube is continuously taken up while being inflated to a predetermined size. Meanwhile, the die is rotated forward or reversed, whereby any possible thickness irregularity can be prevented. The tubular film may be cut at both ends and sealed at one end so that it can be used as a bag. Or, it may be suitably drawn into one continuous film and heat set and/or otherwise treated; thus, as a film having advantageous properties of polybutylene terephthalate resins and having some suitable variations given according to the purpose for which it is used, the product can be offered for various end uses. In the manufacture of films according to the invention, it is possible to adjust film gage in practically same manner as in the conventional inflation process. Generally, film gage is adjustable within the range of 0.005 - 0.1 mm.

In combination with the polybutylene terephthalate resin used in the practice of the invention, various known materials which are usually added to ardinary thermoplastic or thermosetting resins may be suitably used depending upon the performance characteristics required of the product. Materials useful for such purposes include plasticizers, stabilizers, such as antioxidant and U.V. light absorber, antistatic agents,

surface active agents, colorants, such as dyes and pigments, and lubricants and crystallization accelerators (nucleating agents) useful for fluidity improvement. Also, other thermoplastic resins and/or inorganic fillers may be supplementarily used for addition in small amounts insofar as it is not detrimental to the purpose of the invention.

(Advantages of the Invention)

As above described, according to the invention, it is possible to manufacture polybutylene terephthalate resin films by the inflation technique. The invention assures improved productivity and economy as compared with the conventional T-die method, and makes it possible to easily produce even thinner gage films through application of known techniques. Films produced according to the invention have excellent properties inherent to polybutylene terephthalate resins, including, for example, good mechanical, physical, chemical and thermal properties. Further they have a special advantage that they are less subject to moisture and gas permeation. Therefore, they can be advantageously used either alone as such or in lamination with other film or metallic foil for packaging and various other purposes.

(Examples)

The invention will be further illustrated by the following examples. It is understood, however, that the invention is not limited by these examples.

Example 1

A polybutylene terephthalate resin P (IV 2.0, melting point 228°C) was used. From a 50 mmØ extruding machine, with a die diameter of 120mm and a die lip clearance of 1mm, a tubular film was extruded at a resin temperature of 285°C, and a blow ratio of 1.3, and the film was taken off at a take-up rate of 30 m/min while being cooled by air. Thus, a film of 18 μ gage was obtained.

The properties of the film produced were as shown in Table 1.

Property evaluation of the resin and of the film was carried out in the following manner:

IV: measured in orthochlorophenol at 25°C.

Melting point: measured by DSC (heat-up rate 5°C/min) and at a heat absorption peak location.

Tensile strength and elongation: measured according to ASTM D 882

Oxygen and nitrogen permeabilities: measured according to ASTM D 1434.

Table 1

Film gage (μ)	18
Tensile strength (kg/cm^2)	583
Tensile elongation (%)	497
Oxygen permeability (cc/m^2 · day · 1 atm)	293.6
Nitrogen permeability (cc/m^2 · day · 1 atm)	36.4

Examples 2 - 7; Comparative Example 1 - 3

Polybutylene terephthalate resin P, and polybutylene terephthalate resin Q (IV 1.63, melting point 228°C), and polybutylene terephthalate resin R (IV 0.76, melting point 228°C) were used. In an apparatus similar to the one used in Example 1, films of various different gages as shown in Table 2 were produced at varied extrusion resin temperatures. Properties of the films were evaluated. The results are shown in Table 2.

Table 2

IV	Example 2	Example 3	Comp.Exp. 1	Example 4	Example 5	Example 6	Comp.Exp. 2	Comp.Exp. 3
IV	2.0 (Resin P)			1.63 (Resin Q)				0.76 (Resin R)
Extrusion resin temp. (°C)	285	300	315	275	275	285	300	250
Film gage (μ)	35	9	-	35	18	9	-	-
Tensile strength (kg/cm²)	561	833	-	526	534	750	-	-
Tensile elongation (%)	565	470	-	486	415	392	-	-
Oxygen permeability (cc/m²·day·1atm)	151	587	-	160	286	596	-	-
Nitrogen permeability (cc/m²·day·1atm)	18.7	72.7	-	19.0	37.8	73.6	-	-
Moldability	stable	slightly unstable	no good	stable	stable	slightly unstable	no good	no good

Claims

1. A method of manufacturing a polybutylene terephthalate resin film characterised in that a polybutylene terephthalate resin having an inherent viscosity of more than 1.0 is turned into film by an extrusion inflation molding technique within an extrusion resin temperature range defined by the following relation:

Melting point (°C) < extrusion resin temperature (°C) < melting point - 26 + 53 x inherent viscosity (°C)

2. A method of manufacturing a polybutylene terephthalate resin film as set forth in Claim 1 wherein the inherent viscosity of the polybutylene terephthalate resin is 1.2 - 2.5.

3. A method of manufacturing a polybutylene terephthalate resin film as set forth in claim 2, wherein the inherent viscosity of the polybutylene terephthalate resin is 1.5 to 2 2.

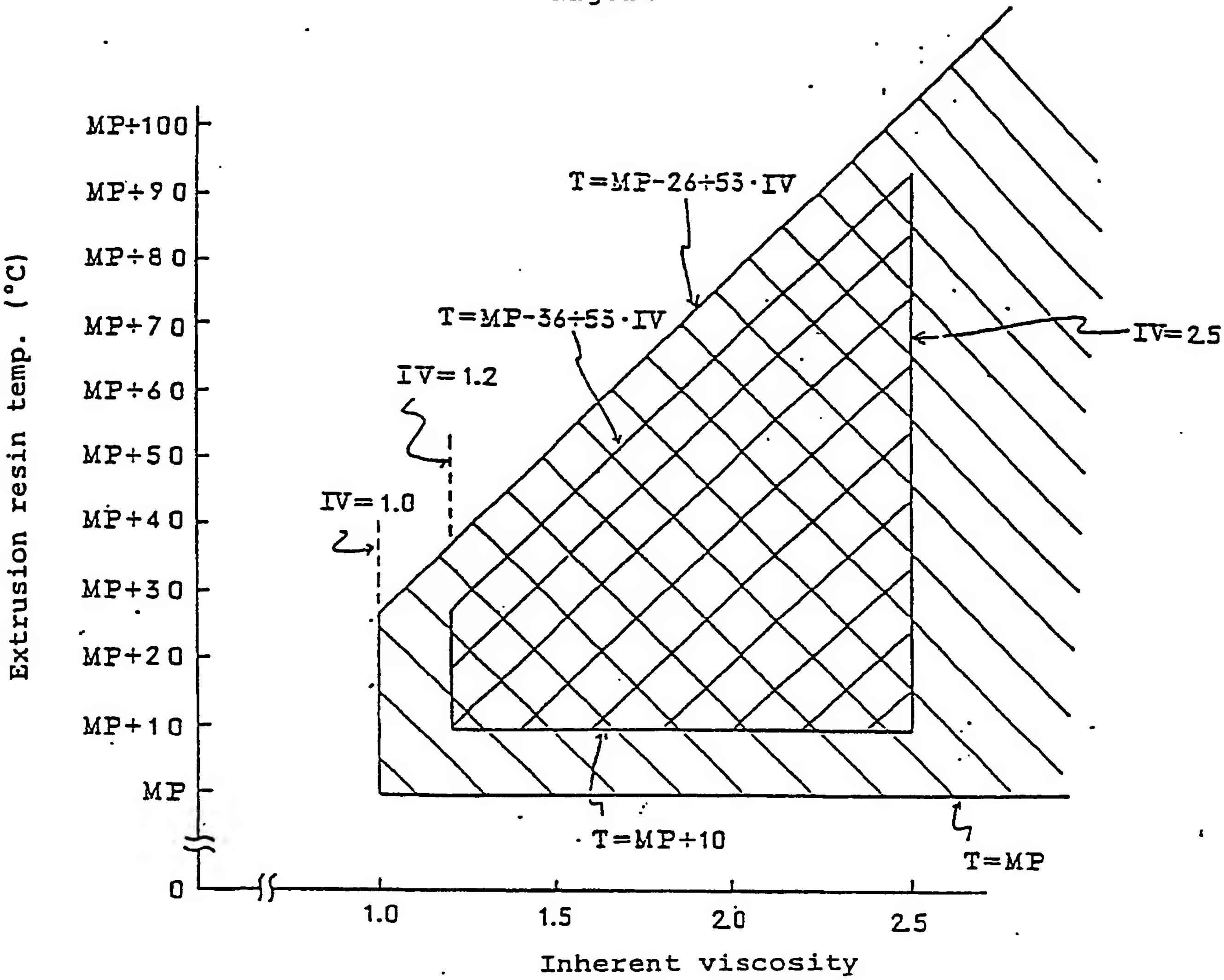
4. A method of manufacturing a polybutylene terephthalate resin film as set forth in any preceding claim, wherein the extrusion resin temperature satisfies the following relation:

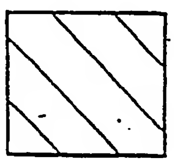
melting point + 10 (°C) ≦ extrusion resin temperature (°C) ≦ melting point - 36 + 53 x inherent viscosity (°C)

5. Polybutylene terephthalate film whenever prepared by a method as claimed in any preceding claim.

6. Polybutylene terephthalate film as set forth in claim 5 and laminated with other film or metallic foil.

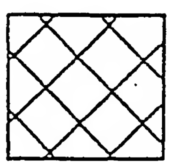
Figure 1





Practicable range: temp. range which satisfies the relation:

MP < T < MP − 26 + 53·IV, 1.0 < IV



Preferred range: temp. range which satisfies the relation:

MP + 10 ≦ T ≦ MP − 36 + 53 · IV, 1.2 ≦ IV ≦ 2.5

where MP: melting point;
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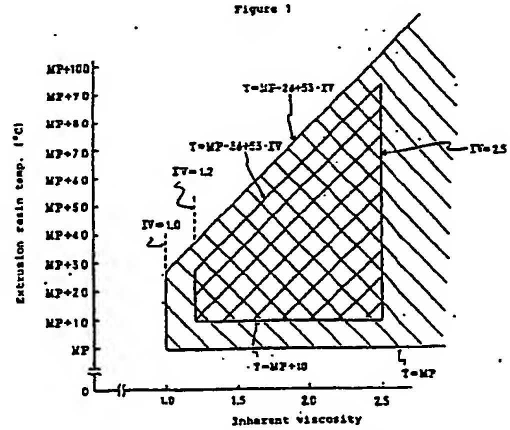

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EUROPEAN SEARCH REPORT

Application Number

EP 88 30 2913

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
Y	EP-A-0 095 995 (THE GOODYEAR TIRE OF RUBBER CY.) * Claims; page 7, lines 9-22; page 8, lines 1-9 * ---	1-5	C 08 J 5/18 B 29 C 47/00
Y	GB-A-1 314 070 (EASTMAN KODAK) * Claims 1,3,16; page 2, lines 44-49; page 3, lines 63-73 * ---	1-6	
A	US-A-4 034 055 (H. STRUTZEL et al.) -----		

TECHNICAL FIELDS SEARCHED (Int. Cl.4)

C 09 J
B 29 C

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	20-02-1990	VAN GOETHEM G.A.J.M.

CATEGORY OF CITED DOCUMENTS

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